

ORIGINAL

Application Based on

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**ORGANIC ELEMENT FOR ELECTROLUMINESCENT DEVICES**

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## **ORGANIC ELEMENT FOR ELECTROLUMINESCENT DEVICES**

### **FIELD OF INVENTION**

This invention relates to organic light emitting diode (OLED)  
5    electroluminescent (EL) device comprising a light-emitting layer containing a  
boron dopant compound containing a bis(azinyl)methene boron group.

### **BACKGROUND OF THE INVENTION**

While organic electroluminescent (EL) devices have been known  
10    for over two decades, their performance limitations have represented a barrier to  
many desirable applications. In simplest form, an organic EL device is comprised  
of an anode for hole injection, a cathode for electron injection, and an organic  
medium sandwiched between these electrodes to support charge recombination  
that yields emission of light. These devices are also commonly referred to as  
15    organic light-emitting diodes, or OLEDs. Representative of earlier organic EL  
devices are Gurnee et al. U.S. Pat. No. 3,172,862, issued Mar. 9, 1965; Gurnee  
U.S. Pat. No. 3,173,050, issued Mar. 9, 1965; Dresner, "Double Injection  
Electroluminescence in Anthracene", RCA Review, Vol. 30, pp. 322-334, 1969;  
and Dresner U.S. Pat. No. 3,710,167, issued Jan. 9, 1973. The organic layers in  
20    these devices, usually composed of a polycyclic aromatic hydrocarbon, were very  
thick (much greater than 1  $\mu\text{m}$ ). Consequently, operating voltages were very high,  
often >100V.

More recent organic EL devices include an organic EL element  
consisting of extremely thin layers (e.g. <1.0  $\mu\text{m}$  ) between the anode and the  
25    cathode. Herein, the organic EL element encompasses the layers between the  
anode and cathode electrodes. Reducing the thickness lowered the resistance of  
the organic layer and has enabled devices that operate at much lower voltage. In a  
basic two-layer EL device structure, described first in US 4,356,429, one organic  
layer of the EL element adjacent to the anode is specifically chosen to transport  
30    holes, therefore, it is referred to as the hole-transporting layer, and the other  
organic layer is specifically chosen to transport electrons, referred to as the  
electron-transporting layer. The interface between the two layers provides an

efficient site for the recombination of the injected hole/electron pair and the resultant electroluminescence.

There have also been proposed three-layer organic EL devices that contain an organic light-emitting layer (LEL) between the hole- transporting layer and electron-transporting layer, such as that disclosed by Tang et al [*J. Applied Physics*, Vol. 65, Pages 3610-3616, 1989]. The light-emitting layer commonly consists of a host material doped with a guest material - dopant , which results in an efficiency improvement and allows color tuning.

Since these early inventions, further improvements in device materials have resulted in improved performance in attributes such as color, stability, luminance efficiency and manufacturability, e.g., as disclosed in US 5,061,569, US 5,409,783, US 5,554,450, US 5,593,788, US 5,683,823, US 5,908,581, US 5,928,802, US 6,020,078, and US 6,208,077, amongst others.

Notwithstanding these developments, there are continuing needs for organic EL device components, such as dopants, that will provide high luminance efficiencies combined with high color purity and long lifetimes.

A useful class of dopants is derived from the 5,6,5-tricyclic pyrromethene-BF<sub>2</sub> complexes and disclosed in US 5,683,823; JP 09,289,081A; and JP 11,097,180A. These materials are characterized by typically narrow emission spectra, which may result in attractively high color purity. However, the green-emitting unsubstituted or alkyl substituted pyrromethene-BF<sub>2</sub> complexes exhibit relatively low quantum efficiencies of electroluminescence. In order to achieve highly efficient OLEDs, one needs to use phenyl rings as substituents thereby extending the conjugated  $\pi$ -system. As a result, the emission wavelength typically becomes red-shifted yielding a reddish amber color, which is the shortest wavelength light that can be emitted by pyrromethene-BF<sub>2</sub> complexes with good efficiency. In simple terms, luminance efficient green OLEDs do not appear to be conveniently obtained with pyrromethene BF<sub>2</sub> complexes used as dopants.

It is a problem to be solved to provide a dopant compound for a light-emitting layer of an OLED device that exhibits improved luminance efficiency.

## SUMMARY OF THE INVENTION

The invention provides an OLED device comprising a light-emitting layer containing a host and a dopant where the dopant comprises a boron compound containing a bis(azinyl)methene boron complex group. The invention 5 also provides a display including such a device and a method of imaging using such a device.

Such a device exhibits improved luminance efficiency.

## BRIEF DESCRIPTION OF THE DRAWINGS

10 FIG. 1 shows a cross-section of a typical OLED device in which this invention may be used.

## DETAILED DESCRIPTION OF THE INVENTION

The invention is generally as described above.

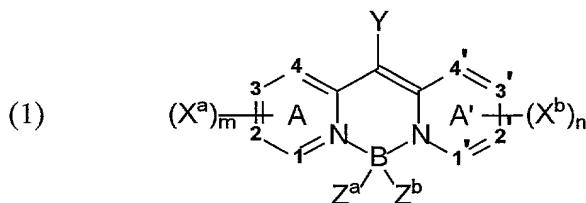
15 An OLED device of the invention is a multilayer electroluminescent device comprising a cathode, an anode, charge-injecting layers (if necessary), charge-transporting layers, and a light-emitting layer (LEL) comprising a host and at least one dopant, a bis(azinyl)methene boron compound. The term azine or azinyl refers to a six-membered aromatic ring system containing 20 at least one nitrogen as defined by the Hantzsch-Widman stems [*The Naming and Indexing of Chemical Substances for Chemical Abstracts-A Reprint of Index IV (Chemical Substance Index Names) from the Chemical Abstracts – 1992 Index Guide*; American Chemical Society: Columbus, OH, 1992; paragraph 146].

25 Suitably, the light-emitting layer of the device comprises a host and dopant where the dopant is present in an amount of up to 10 wt % of the host, more typically from 0.1-5.0 wt % of the host. The group is suitably a 6,6,6-tricyclic bis(azinyl)methene boron complex group, and usefully a bis(pyridinyl)methene boron complex group. Good results are obtained when at least one or both of the pyridyl groups are substituted, particularly where at least 30 one or both of the pyridyl groups has substituent groups joined to form a fused ring.

The benefit imparted by the dopant does not appear to be host specific. Desirable hosts include those based on a chelated oxinoid compound or an anthracene compound. Particular examples of hosts are tris(8-quinolinolato)aluminum (III) and 2-*tert*-butyl-9,10-di-(2-naphthyl)anthracene.

5 Embodiments of the dopants useful in the invention provide an emitted light having a green hue. Substituents are selected to provide embodiments that exhibit a reduced loss of initial luminance compared to the device containing no boron compound of claim 1.

10 Compounds useful in the invention are suitably represented by Formula (1):



wherein

15 A and A' represent independent azine ring systems corresponding to 6-membered aromatic ring systems containing at least one nitrogen;

each X<sup>a</sup> and X<sup>b</sup> is an independently selected substituent, two of which may join to form a fused ring to A or A';

m and n are independently 0 to 4;

20 Y is H or a substituent;

Z<sup>a</sup> and Z<sup>b</sup> are independently selected substituents; and

1, 2, 3, 4, 1', 2', 3', and 4' are independently selected as either carbon or nitrogen atoms.

In the device, 1, 2, 3, 4, 1', 2', 3', and 4' are conveniently all carbon atoms. The device may desirably contain at least one or both of ring A or A' that contains substituents joined to form a fused ring. In one useful embodiment, there is present at least one X<sup>a</sup> or X<sup>b</sup> group selected from the group consisting of halide and alkyl, aryl, alkoxy, and aryloxy groups. In another embodiment, there is

present a  $Z^a$  and  $Z^b$  group are independently selected from the group consisting of fluorine and alkyl, aryl, alkoxy and aryloxy groups. A desirable embodiment is where  $Z^a$  and  $Z^b$  are F. Y is suitably hydrogen or a substituent such as an alkyl, aryl, or heterocyclic group.

5 The emission wavelength of these compounds may be adjusted to some extent by appropriate substitution around the central bis(azinyl)methene boron group to meet a color aim, namely green.

	Ring System	Illustrative Example	Structure
Inventive	6,6,6	Pyridomethene $BF_2$ complex	
Comparative	5,6,5	Pyrromethene $BF_2$ complex	

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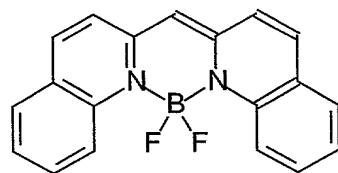
The bis(azinyl)methene boron compound is usually doped into a host compound, which represents the light-emitting layer between the hole-transporting and electron-transporting layers. The host is chosen such that there is efficient energy transfer from the host to the bis(azinyl)methene boron compound.

15 The bis(azinyl)methene boron complex emits from the excited state to afford a bright, highly-efficient, stable EL device.

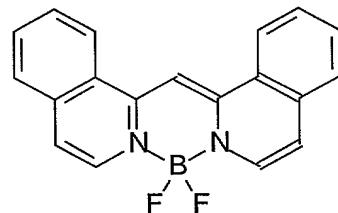
The EL device of the invention is useful in any device where light emission is desired such as a lamp or a component in a static or motion imaging device, such as a television, cell phone, DVD player, or computer monitor.

20 Illustrative examples of bis(azinyl)methene boron compounds useful in the present invention are the following:

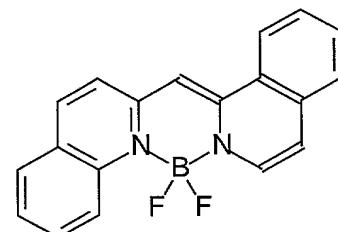
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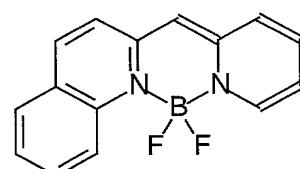
Inv-2



Inv-3



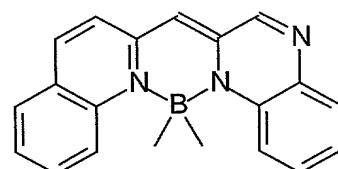
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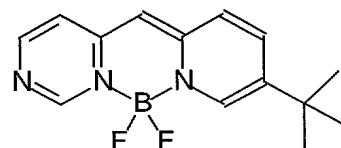
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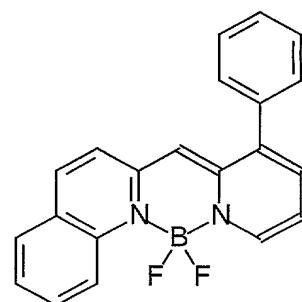
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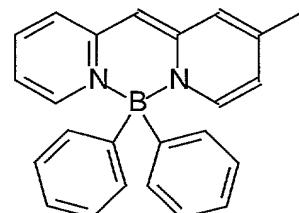
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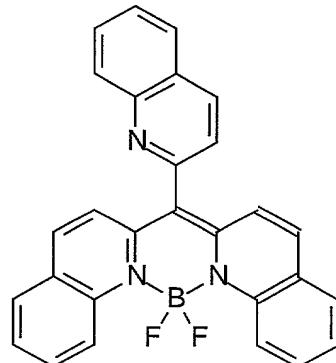
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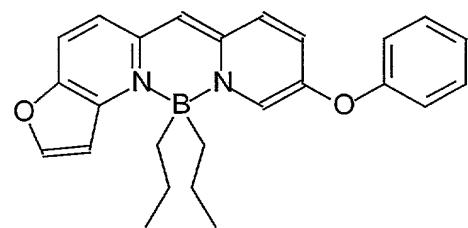
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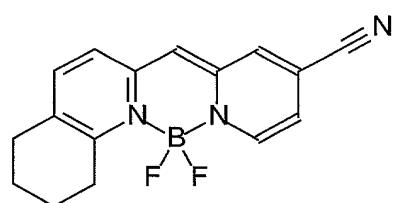
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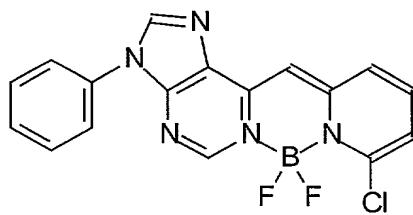
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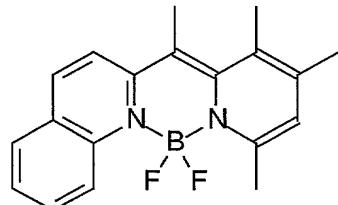
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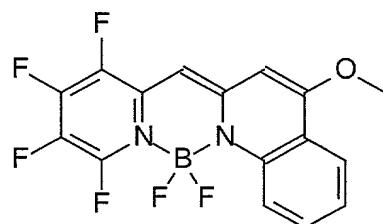
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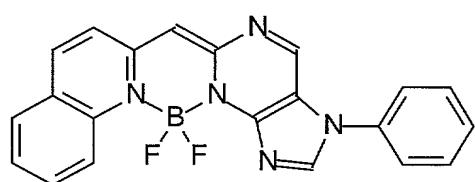
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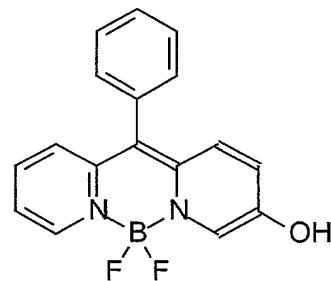
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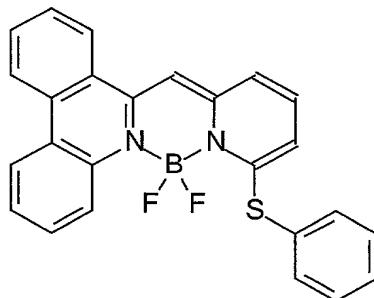
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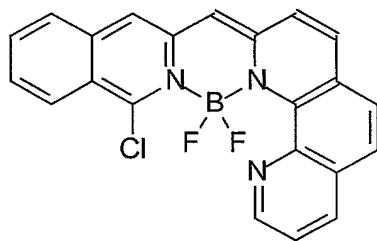
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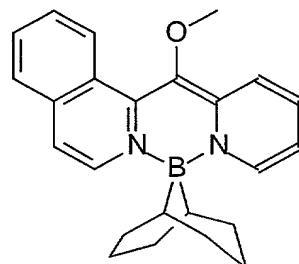
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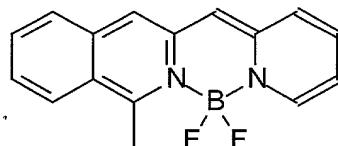
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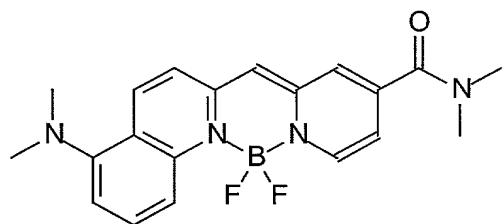
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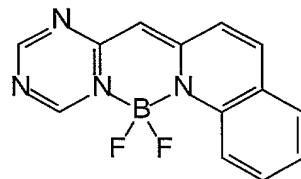
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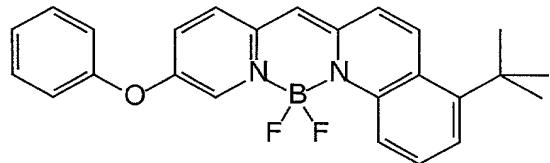
Inv-22



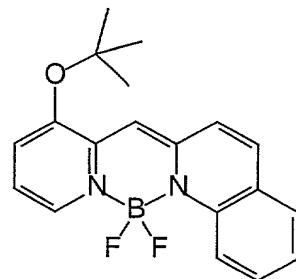
Inv-23



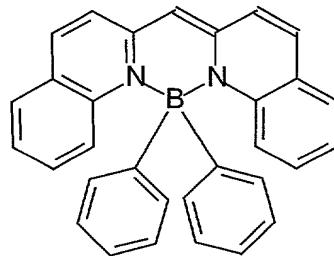
Inv-24



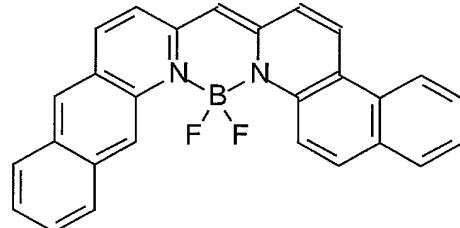
Inv-25



Inv-26



Inv-27



Embodiments of the invention provide not only improved  
luminance efficiency but also improved stability under load and a more desirable  
green hue as evidenced by the location and shape of the absorption curve of the  
5 emitted light.

Unless otherwise specifically stated, use of the term "substituted"  
or "substituent" means any group or atom other than hydrogen (what about  
deuterium). Additionally, when the term "group" is used, it means that when a  
substituent group contains a substitutable hydrogen, it is also intended to  
10 encompass not only the substituent's unsubstituted form, but also its form further  
substituted with any substituent group or groups as herein mentioned, so long as  
the substituent does not destroy properties necessary for device utility. Suitably, a  
substituent group may be halogen or may be bonded to the remainder of the  
molecule by an atom of carbon, silicon, oxygen, nitrogen, phosphorous, sulfur,  
15 selenium, or boron. The substituent may be, for example, halogen, such as chloro,

bromo or fluoro; nitro; hydroxyl; cyano; carboxyl; or groups which may be further substituted, such as alkyl, including straight or branched chain or cyclic alkyl, such as methyl, trifluoromethyl, ethyl, *t*-butyl, 3-(2,4-di-*t*-pentylphenoxy) propyl, and tetradecyl; alkenyl, such as ethylene, 2-butene; alkoxy, such as methoxy, 5 ethoxy, propoxy, butoxy, 2-methoxyethoxy, *sec*-butoxy, hexyloxy, 2-ethylhexyloxy, tetradecyloxy, 2-(2,4-di-*t*-pentylphenoxy)ethoxy, and 2-dodecyloxyethoxy; aryl such as phenyl, 4-*t*-butylphenyl, 2,4,6-trimethylphenyl, naphthyl; aryloxy, such as phenoxy, 2-methylphenoxy, alpha- or beta-naphthoxy, and 4-tolyloxy; carbonamido, such as acetamido, benzamido, 10 butyramido, tetradecanamido, alpha-(2,4-di-*t*-pentyl-phenoxy)acetamido, alpha-(2,4-di-*t*-pentylphenoxy)butyramido, alpha-(3-pentadecylphenoxy)-hexanamido, alpha-(4-hydroxy-3-*t*-butylphenoxy)-tetradecanamido, 2-oxo-pyrrolidin-1-yl, 2-oxo-5-tetradecylpyrrolin-1-yl, *N*-methyltetradecanamido, *N*-succinimido, *N*-phthalimido, 2,5-dioxo-1-oxazolidinyl, 3-dodecyl-2,5-dioxo-1-imidazolyl, and *N*-15 acetyl-*N*-dodecylamino, ethoxycarbonylamino, phenoxy carbonylamino, benzyloxycarbonylamino, hexadecyloxycarbonylamino, 2,4-di-*t*-butylphenoxy carbonylamino, phenylcarbonylamino, 2,5-(di-*t*-pentylphenyl)carbonylamino, *p*-dodecyl-phenylcarbonylamino, *p*-tolylcarbonylamino, *N*-methylureido, *N,N*-dimethylureido, *N*-methyl-*N*-20 dodecylureido, *N*-hexadecylureido, *N,N*-dioctadecylureido, *N,N*-dioctyl-*N'*-ethylureido, *N*-phenylureido, *N,N*-diphenylureido, *N*-phenyl-*N*-*p*-tolylureido, *N*-(*m*-hexadecylphenyl)ureido, *N,N*-(2,5-di-*t*-pentylphenyl)-*N'*-ethylureido, and *t*-butylcarbonamido; sulfonamido, such as methylsulfonamido, benzenesulfonamido, *p*-tolylsulfonamido, *p*-dodecylbenzenesulfonamido, *N*-25 methyltetradecylsulfonamido, *N,N*-dipropyl-sulfamoylamino, and hexadecylsulfonamido; sulfamoyl, such as *N*-methylsulfamoyl, *N*-ethylsulfamoyl, *N,N*-dipropylsulfamoyl, *N*-hexadecylsulfamoyl, *N,N*-dimethylsulfamoyl, *N*-[3-(dodecyloxy)propyl]sulfamoyl, *N*-[4-(2,4-di-*t*-pentylphenoxy)butyl]sulfamoyl, *N*-methyl-*N*-tetradecylsulfamoyl, and *N*-dodecylsulfamoyl; carbamoyl, such as *N*-30 methylcarbamoyl, *N,N*-dibutylcarbamoyl, *N*-octadecylcarbamoyl, *N*-[4-(2,4-di-*t*-pentylphenoxy)butyl]carbamoyl, *N*-methyl-*N*-tetradecylcarbamoyl, and *N,N*-

dioctylcarbamoyl; acyl, such as acetyl, (2,4-di-*t*-amylphenoxy)acetyl, phenoxy carbonyl, *p*-dodecyloxyphenoxy carbonyl methoxycarbonyl, butoxycarbonyl, tetradecyloxycarbonyl, ethoxycarbonyl, benzyloxycarbonyl, 3-pentadecyloxycarbonyl, and dodecyloxycarbonyl; sulfonyl, such as

5 methoxysulfonyl, octyloxysulfonyl, tetradecyloxysulfonyl, 2-ethylhexyloxysulfonyl, phenoxy sulfonyl, 2,4-di-*t*-pentylphenoxy sulfonyl, methylsulfonyl, octylsulfonyl, 2-ethylhexylsulfonyl, dodecylsulfonyl, hexadecylsulfonyl, phenylsulfonyl, 4-nonylphenylsulfonyl, and *p*-tolylsulfonyl; sulfonyloxy, such as dodecylsulfonyloxy, and hexadecylsulfonyloxy; sulfinyl,

10 such as methylsulfinyl, octylsulfinyl, 2-ethylhexylsulfinyl, dodecylsulfinyl, hexadecylsulfinyl, phenylsulfinyl, 4-nonylphenylsulfinyl, and *p*-tolylsulfinyl; thio, such as ethylthio, octylthio, benzylthio, tetradecylthio, 2-(2,4-di-*t*-pentylphenoxy)ethylthio, phenylthio, 2-butoxy-5-*t*-octylphenylthio, and *p*-tolylthio; acyloxy, such as acetoxy, benzoxy, octadecanoyloxy, *p*-

15 dodecylamidobenzoyloxy, *N*-phenylcarbamoyloxy, *N*-ethylcarbamoyloxy, and cyclohexylcarbonyloxy; amine, such as phenylanilino, 2-chloroanilino, diethylamine, dodecylamine; imino, such as 1 (*N*-phenylimido)ethyl, *N*-succinimido or 3-benzylhydantoinyl; phosphate, such as dimethylphosphate and ethylbutylphosphate; phosphite, such as diethyl and dihexylphosphite; a

20 heterocyclic group, a heterocyclic oxy group or a heterocyclic thio group, each of which may be substituted and which contain a 3 to 7 membered heterocyclic ring composed of carbon atoms and at least one hetero atom selected from the group consisting of oxygen, nitrogen, sulfur, phosphorous, or boron. such as 2-furyl, 2-thienyl, 2-benzimidazolyloxy or 2-benzothiazolyl; quaternary ammonium, such as triethylammonium; quaternary phosphonium, such as triphenylphosphonium; and silyloxy, such as trimethylsilyloxy.

25

If desired, the substituents may themselves be further substituted one or more times with the described substituent groups. The particular substituents used may be selected by those skilled in the art to attain the desired desirable properties

30 for a specific application and can include, for example, electron-withdrawing groups, electron-donating groups, and steric groups. When a molecule may have two or more substituents, the substituents may be joined together to form a ring

such as a fused ring unless otherwise provided. Generally, the above groups and substituents thereof may include those having up to 48 carbon atoms, typically 1 to 36 carbon atoms and usually less than 24 carbon atoms, but greater numbers are possible depending on the particular substituents selected.

5

#### General device architecture

The present invention can be employed in most OLED device configurations. These include very simple structures comprising a single anode and cathode to more complex devices, such as passive matrix displays comprised of orthogonal arrays of anodes and cathodes to form pixels, and active-matrix displays where each pixel is controlled independently, for example, with a thin film transistor (TFT).

There are numerous configurations of the organic layers wherein the present invention can be successfully practiced. Essential requirements are a cathode, an anode, an HTL and an LEL. A more typical structure is shown in Figure 1 and contains a substrate **101**, an anode **103**, an optional hole-injecting layer **105**, a hole-transporting layer **107**, a light-emitting layer **109**, an electron-transporting layer **111**, and a cathode **113**. These layers are described in detail below. Note that the substrate may alternatively be located adjacent to the cathode, or the substrate may actually constitute the anode or cathode. Also, the total combined thickness of the organic layers is preferably less than 500 nm.

#### Substrate

The substrate **101** can either be light transmissive or opaque, depending on the intended direction of light emission. The light transmissive property is desirable for viewing the EL emission through the substrate. Transparent glass or organic material are commonly employed in such cases. For applications where the EL emission is viewed through the top electrode, the transmissive characteristic of the bottom support is immaterial, and therefore can be light transmissive, light absorbing or light reflective. Substrates for use in this case include, but are not limited to, glass, plastic, semiconductor materials,

ceramics, and circuit board materials. Of course it is necessary to provide in these device configurations a light-transparent top electrode.

Anode

5 The conductive anode layer **103** is commonly formed over the substrate and, when EL emission is viewed through the anode, should be transparent or substantially transparent to the emission of interest. Common transparent anode materials used in this invention are indium-tin oxide (ITO) and tin oxide, but other metal oxides can work including, but not limited to,  
10 aluminum- or indium-doped zinc oxide (IZO), magnesium-indium oxide, and nickel-tungsten oxide. In addition to these oxides, metal nitrides, such as gallium nitride, and metal selenides, such as zinc selenide, and metal sulfides, such as zinc sulfide, can be used in layer **103**. For applications where EL emission is viewed through the top electrode, the transmissive characteristics of layer **103** are  
15 immaterial and any conductive material can be used, transparent, opaque or reflective. Example conductors for this application include, but are not limited to, gold, iridium, molybdenum, palladium, and platinum. Typical anode materials, transmissive or otherwise, have a work function of 4.1 eV or greater. Desired anode materials are commonly deposited by any suitable means such as  
20 evaporation, sputtering, chemical vapor deposition, or electrochemical means. Anodes can be patterned using well-known photolithographic processes.

Hole-Injecting Layer (HIL)

While not always necessary, it is often useful that a hole-injecting layer **105** be provided between anode **103** and hole-transporting layer **107**. The hole-injecting material can serve to improve the film formation property of subsequent organic layers and to facilitate injection of holes into the hole-transporting layer. Suitable materials for use in the hole-injecting layer include, but are not limited to, porphyrinic compounds such as those described in US  
25 4,720,432, and plasma-deposited fluorocarbon polymers such as those described in US 6,208,075. Alternative hole-injecting materials reportedly useful in organic  
30 EL devices are described in EP 0 891 121 A1 and EP 1 029 909 A1.

Hole-Transporting Layer (HTL)

The hole-transporting layer 107 of the organic EL device contains at least one hole-transporting compound such as an aromatic tertiary amine, where the latter is understood to be a compound containing at least one trivalent nitrogen atom that is bonded only to carbon atoms, at least one of which is a member of an aromatic ring. In one form the aromatic tertiary amine can be an arylamine, such as a monoarylamine, diarylamine, triarylamine, or a polymeric arylamine group.

Exemplary monomeric triarylamines are illustrated by Klupfel et al. US

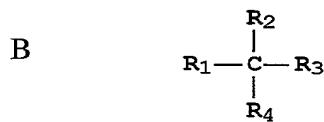
10 3,180,730. Other suitable triarylamines substituted with one or more vinyl radicals and/or comprising at least one active hydrogen containing group are disclosed by Brantley et al US 3,567,450 and US 3,658,520.

A more preferred class of aromatic tertiary amines are those which include at least two aromatic tertiary amine moieties as described in US 4,720,432 and US 5,061,569. Such compounds include those represented by structural formula (A).



wherein Q<sub>1</sub> and Q<sub>2</sub> are independently selected aromatic tertiary amine moieties and G is a linking group such as an arylene, cycloalkylene, or alkylene group of a carbon to carbon bond. In one embodiment, at least one of Q<sub>1</sub> or Q<sub>2</sub> contains a polycyclic fused ring group, e.g., a naphthalene. When G is an aryl group, it is conveniently a phenylene, biphenylene, or naphthalene group.

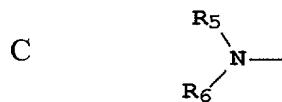
A useful class of triarylamine groups satisfying structural formula (A) and containing two triarylamine groups is represented by structural formula (B):



where

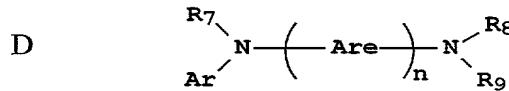
R<sub>1</sub> and R<sub>2</sub> each independently represents a hydrogen atom, an aryl group, or an alkyl group or R<sub>1</sub> and R<sub>2</sub> together represent the atoms completing a cycloalkyl group; and

5 R<sub>3</sub> and R<sub>4</sub> each independently represents an aryl group, which is in turn substituted with a diaryl substituted amino group, as indicated by structural formula (C):



10 wherein R<sub>5</sub> and R<sub>6</sub> are independently selected aryl groups. In one embodiment, at least one of R<sub>5</sub> or R<sub>6</sub> contains a polycyclic fused ring group, e.g., a naphthalene.

Another class of aromatic tertiary amine groups are the tetraaryldiamines. Desirable tetraaryldiamines groups include two diarylamino groups, such as indicated by formula (C), linked through an arylene group. Useful 15 tetraaryldiamines include those represented by formula (D).



wherein

each Are is an independently selected arylene group, such as a phenylene or anthracene group,

20 n is an integer of from 1 to 4, and

Ar, R<sub>7</sub>, R<sub>8</sub>, and R<sub>9</sub> are independently selected aryl groups.

In a typical embodiment, at least one of Ar, R<sub>7</sub>, R<sub>8</sub>, and R<sub>9</sub> is a polycyclic fused ring group, e.g., a naphthalene

25 The various alkyl, alkylene, aryl, and arylene groups of the foregoing structural formulae (A), (B), (C), (D), can each in turn be substituted. Typical substituents include alkyl groups, alkoxy groups, aryl groups, aryloxy groups, and halogen such as fluoride, chloride, and bromide. The various alkyl

and alkylene groups typically contain from about 1 to 6 carbon atoms. The cycloalkyl moieties can contain from 3 to about 10 carbon atoms, but typically contain five, six, or seven ring carbon atoms--e.g., cyclopentyl, cyclohexyl, and cycloheptyl ring structures. The aryl and arylene groups are usually phenyl and phenylene moieties.

The hole-transporting layer can be formed of a single or a mixture of aromatic tertiary amine compounds. Specifically, one may employ a triarylamine, such as a triarylamine satisfying the formula (B), in combination with a tetraaryldiamine, such as indicated by formula (D). When a triarylamine is employed in combination with a tetraaryldiamine, the latter is positioned as a layer interposed between the triarylamine and the electron injecting and transporting layer. Illustrative of useful aromatic tertiary amines are the following:

1,1-Bis(4-di-*p*-tolylaminophenyl)cyclohexane  
15 1,1-Bis(4-di-*p*-tolylaminophenyl)-4-phenylcyclohexane  
4,4'-Bis(diphenylamino)quadriphenyl  
Bis(4-dimethylamino-2-methylphenyl)-phenylmethane  
*N,N,N-Tri(p-tolyl)amine*  
4-(di-*p*-tolylamino)-4'-[4(di-*p*-tolylamino)-styryl]stilbene  
20 *N,N,N',N'-Tetra-p-tolyl-4-4'-diaminobiphenyl*  
*N,N,N',N'-Tetraphenyl-4,4'-diaminobiphenyl*  
*N,N,N',N'-tetra-1-naphthyl-4,4'-diaminobiphenyl*  
*N,N,N',N'-tetra-2-naphthyl-4,4'-diaminobiphenyl*  
*N-Phenylcarbazole*  
25 4,4'-Bis[*N*-(1-naphthyl)-*N*-phenylamino]biphenyl  
4,4'-Bis[*N*-(1-naphthyl)-*N*-(2-naphthyl)amino]biphenyl  
4,4"-Bis[*N*-(1-naphthyl)-*N*-phenylamino]p-terphenyl  
4,4'-Bis[*N*-(2-naphthyl)-*N*-phenylamino]biphenyl  
4,4'-Bis[*N*-(3-acenaphthyl)-*N*-phenylamino]biphenyl  
30 1,5-Bis[*N*-(1-naphthyl)-*N*-phenylamino]naphthalene  
4,4'-Bis[*N*-(9-anthryl)-*N*-phenylamino]biphenyl  
4,4"-Bis[*N*-(1-anthryl)-*N*-phenylamino]-*p*-terphenyl

4,4'-Bis[N-(2-phenanthryl)-N-phenylamino]biphenyl  
4,4'-Bis[N-(8-fluoranthenyl)-N-phenylamino]biphenyl  
4,4'-Bis[N-(2-pyrenyl)-N-phenylamino]biphenyl  
4,4'-Bis[N-(2-naphthacenyl)-N-phenylamino]biphenyl  
5 4,4'-Bis[N-(2-perylenyl)-N-phenylamino]biphenyl  
4,4'-Bis[N-(1-coronenyl)-N-phenylamino]biphenyl  
2,6-Bis(di-*p*-tolylamino)naphthalene  
2,6-Bis[di-(1-naphthyl)amino]naphthalene  
2,6-Bis[N-(1-naphthyl)-N-(2-naphthyl)amino]naphthalene  
10 *N,N,N',N'*-Tetra(2-naphthyl)-4,4"-diamino-*p*-terphenyl  
4,4'-Bis{N-phenyl-N-[4-(1-naphthyl)-phenyl]amino}biphenyl  
4,4'-Bis[N-phenyl-N-(2-pyrenyl)amino]biphenyl  
2,6-Bis[N,N-di(2-naphthyl)amine]fluorine  
1,5-Bis[N-(1-naphthyl)-N-phenylamino]naphthalene  
15

Another class of useful hole-transporting materials includes polycyclic aromatic compounds as described in EP 1 009 041. In addition, polymeric hole-transporting materials can be used such as poly(*N*-vinylcarbazole) (PVK), polythiophenes, polypyrrole, polyaniline, and copolymers such as 20 poly(3,4-ethylenedioxythiophene) / poly(4-styrenesulfonate) also called PEDOT/PSS.

#### Light-Emitting Layer (LEL)

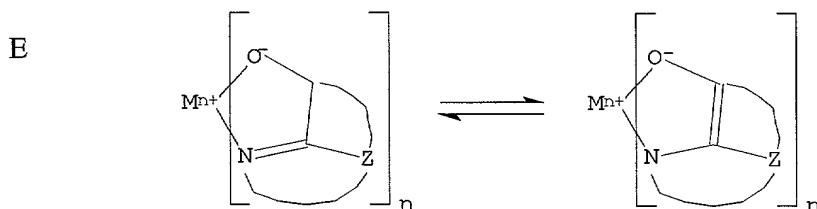
As more fully described in US 4,769,292 and 5,935,721, the light-emitting layer (LEL) **109** of the organic EL element comprises a luminescent or fluorescent material where electroluminescence is produced as a result of electron-hole pair recombination in this region. The light-emitting layer can be comprised of a single material, but more commonly consists of a host material doped with a guest compound or compounds where light emission comes primarily from the 25 guest compound. The host materials in the light-emitting layer can be an electron-transporting material, as defined below, a hole-transporting material, as defined above, or another material or combination of materials that 30

support hole-electron recombination. The dopant is usually chosen from highly fluorescent dyes, but phosphorescent compounds, e.g., transition metal complexes as described in WO 98/55561, WO 00/18851, WO 00/57676, and WO 00/70655 are also useful. Dopants are typically coated as 0.01 to 10 % by weight into the host material.

5 An important relationship for choosing a dye as a dopant is a comparison of the bandgap potential which is defined as the energy difference between the highest occupied molecular orbital and the lowest unoccupied molecular orbital of the molecule. For efficient energy transfer from the host to 10 the dopant molecule, a necessary condition is that the band gap of the dopant is smaller than that of the host material.

15 Host and emitting molecules known to be of use include, but are not limited to, those disclosed in US 4,768,292, US 5,141,671, US 5,150,006, US 5,151,629, US 5,405,709, US 5,484,922, US 5,593,788, US 5,645,948, US 5,683,823, US 5,755,999, US 5,928,802, US 5,935,720, US 5,935,721, and US 6,020,078.

20 Metal complexes of 8-hydroxyquinoline and similar derivatives (Formula E) constitute one class of useful host compounds capable of supporting electroluminescence, and are particularly suitable for light emission of wavelengths longer than 500 nm, e.g., green, yellow, orange, and red.



wherein

M represents a metal;

n is an integer of from 1 to 4; and

25 Z independently in each occurrence represents the atoms completing a nucleus having at least two fused aromatic rings.

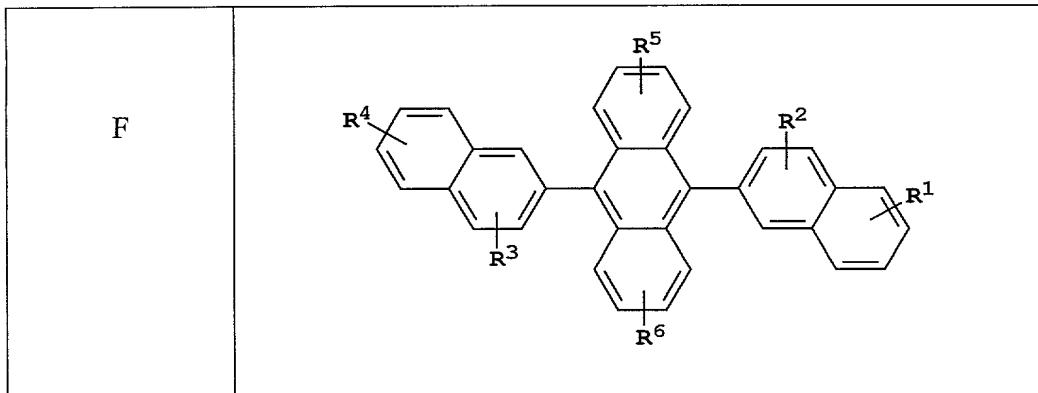
From the foregoing it is apparent that the metal can be monovalent, divalent, trivalent, or tetravalent metal. The metal can, for example, be an alkali metal, such as lithium, sodium, or potassium; an alkaline earth metal, such as magnesium or calcium; an earth metal, such aluminum or gallium, or a transition metal such as zinc or zirconium. Generally any monovalent, divalent, trivalent, or tetravalent metal known to be a useful chelating metal can be employed.

Z completes a heterocyclic nucleus containing at least two fused aromatic rings, at least one of which is an azole or azine ring. Additional rings, including both aliphatic and aromatic rings, can be fused with the two required rings, if required. To avoid adding molecular bulk without improving on function the number of ring atoms is usually maintained at 18 or less.

Illustrative of useful chelated oxinoid compounds are the following:

- CO-1: Aluminum trisoxine [alias, tris(8-quinolinolato)aluminum(III)]
- 15 CO-2: Magnesium bisoxine [alias, bis(8-quinolinolato)magnesium(II)]
- CO-3: Bis[benzo{f}-8-quinolinolato]zinc (II)
- CO-4: Bis(2-methyl-8-quinolinolato)aluminum(III)- $\mu$ -oxo-bis(2-methyl-8-quinolinolato) aluminum(III)
- CO-5: Indium trisoxine [alias, tris(8-quinolinolato)indium]
- 20 CO-6: Aluminum tris(5-methyloxine) [alias, tris(5-methyl-8-quinolinolato) aluminum(III)]
- CO-7: Lithium oxine [alias, (8-quinolinolato)lithium(I)]
- CO-8: Gallium oxine [alias, tris(8-quinolinolato)gallium(III)]
- CO-9: Zirconium oxine [alias, tetra(8-quinolinolato)zirconium(IV)]
- 25 CO-10: Bis(2-methyl-8-quinolinato)-4-phenylphenolatoaluminum (III)

Derivatives of 9,10-di-(2-naphthyl)anthracene (Formula F) constitute one class of useful hosts capable of supporting electroluminescence, and are particularly suitable for light emission of wavelengths longer than 400 nm, 30 e.g., blue, green, yellow, orange or red.



wherein:  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^5$ , and  $R^6$  represent hydrogen or one or more substituents selected from the following groups:

Group 1: hydrogen, alkyl and alkoxy groups typically having from 1 to 24  
5 carbon atoms;

Group 2: a ring group, typically having from 6 to 20 carbon atoms;

Group 3: the atoms necessary to complete a carbocyclic fused ring group such as naphthyl, anthracenyl, pyrenyl, and perylenyl groups, typically having from 6 to 30 carbon atoms;

10 Group 4: the atoms necessary to complete a heterocyclic fused ring group such as furyl, thienyl, pyridyl, and quinolinyl groups, typically having from 5 to 24 carbon atoms;

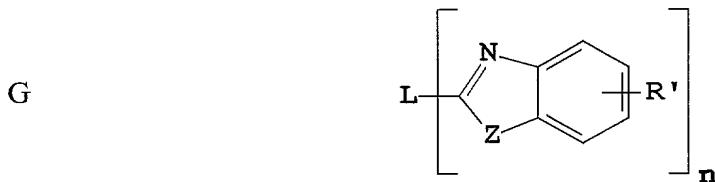
Group 5: an alkoxyamino, alkylamino, and arylamino group typically having from 1 to 24 carbon atoms; and

15 Group 6: fluorine, chlorine, bromine and cyano radicals.

Illustrative examples include 9,10-di-(2-naphthyl)anthracene and 2-  
t-butyl-9,10-di-(2-naphthyl)anthracene. Other anthracene derivatives can be  
useful as a host in the LEL, including derivatives of 9,10-bis[4-(2,2-  
20 diphenylethenyl)phenyl]anthracene, and phenylanthracene derivatives as  
described in EP 681,019.

Benzazole derivatives (Formula G) constitute another class of useful hosts capable of supporting electroluminescence, and are particularly

suitable for light emission of wavelengths longer than 400 nm, e.g., blue, green, yellow, orange or red.



where:

n is an integer of 3 to 8;

5 Z is -O, -NR or -S where R is H or a substituent; and

R' represents one or more optional substituents where R and each R' are H or alkyl groups such as propyl, t-butyl, and heptyl groups typically having from 1 to 24 carbon atoms; carbocyclic or heterocyclic ring groups such as phenyl and naphthyl, furyl, thienyl, pyridyl, and quinolinyl groups and atoms necessary to

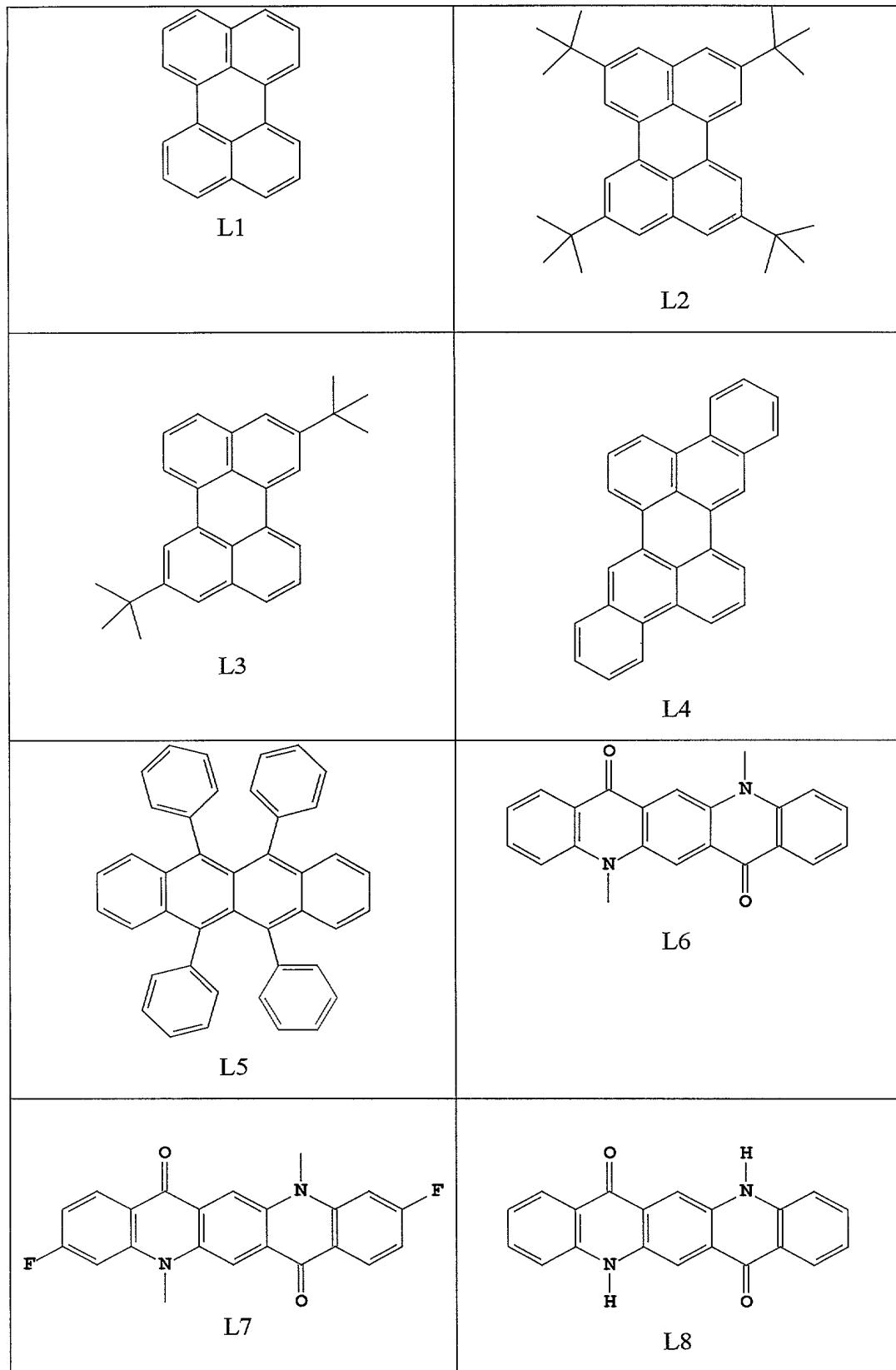
10 complete a fused aromatic ring group typically having from 5 to 20 carbon atoms; and halo such as chloro, and fluoro;

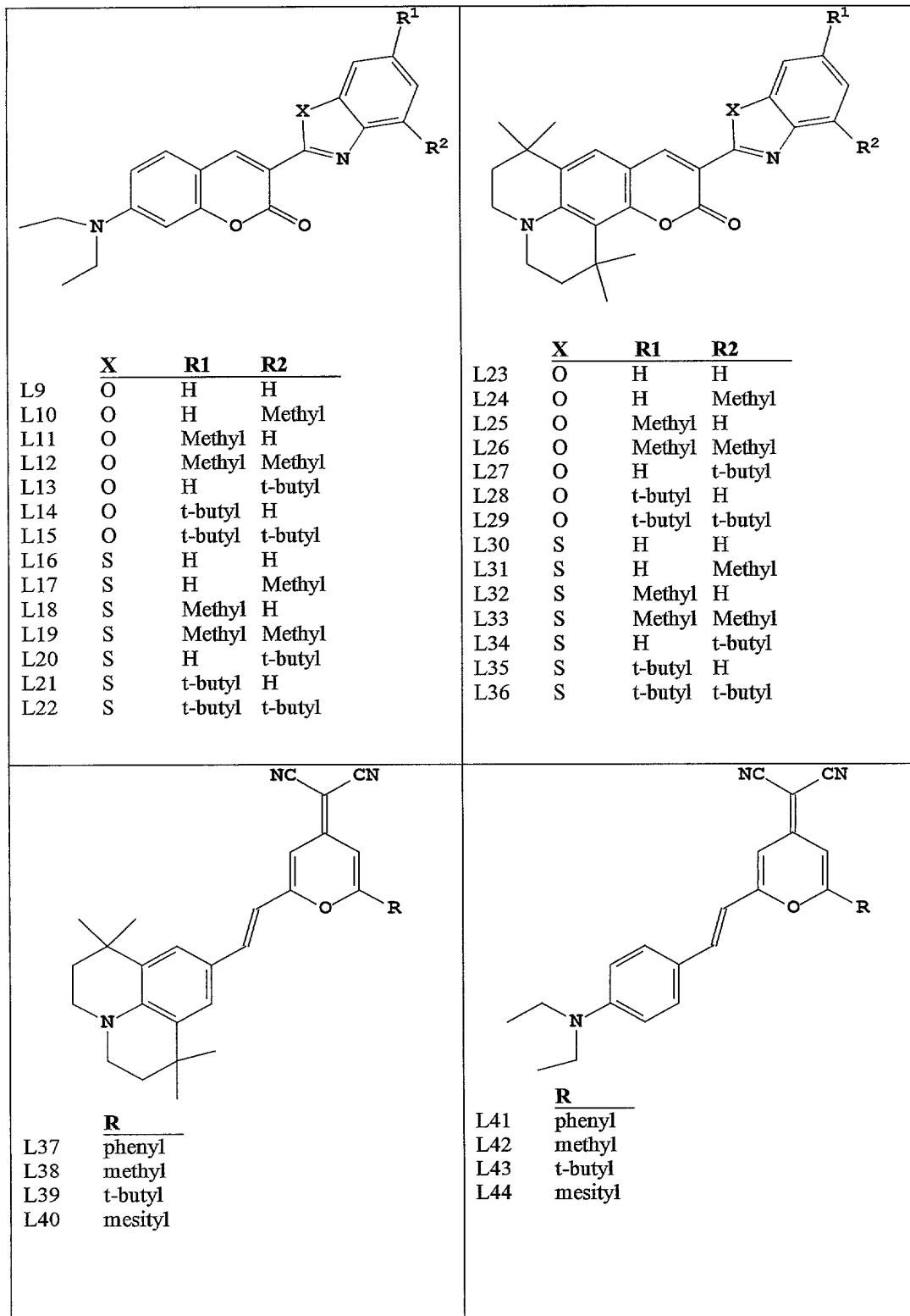
L is a linkage unit usually comprising an alkyl or ary group which conjugately or unconjugately connects the multiple benzazoles together.

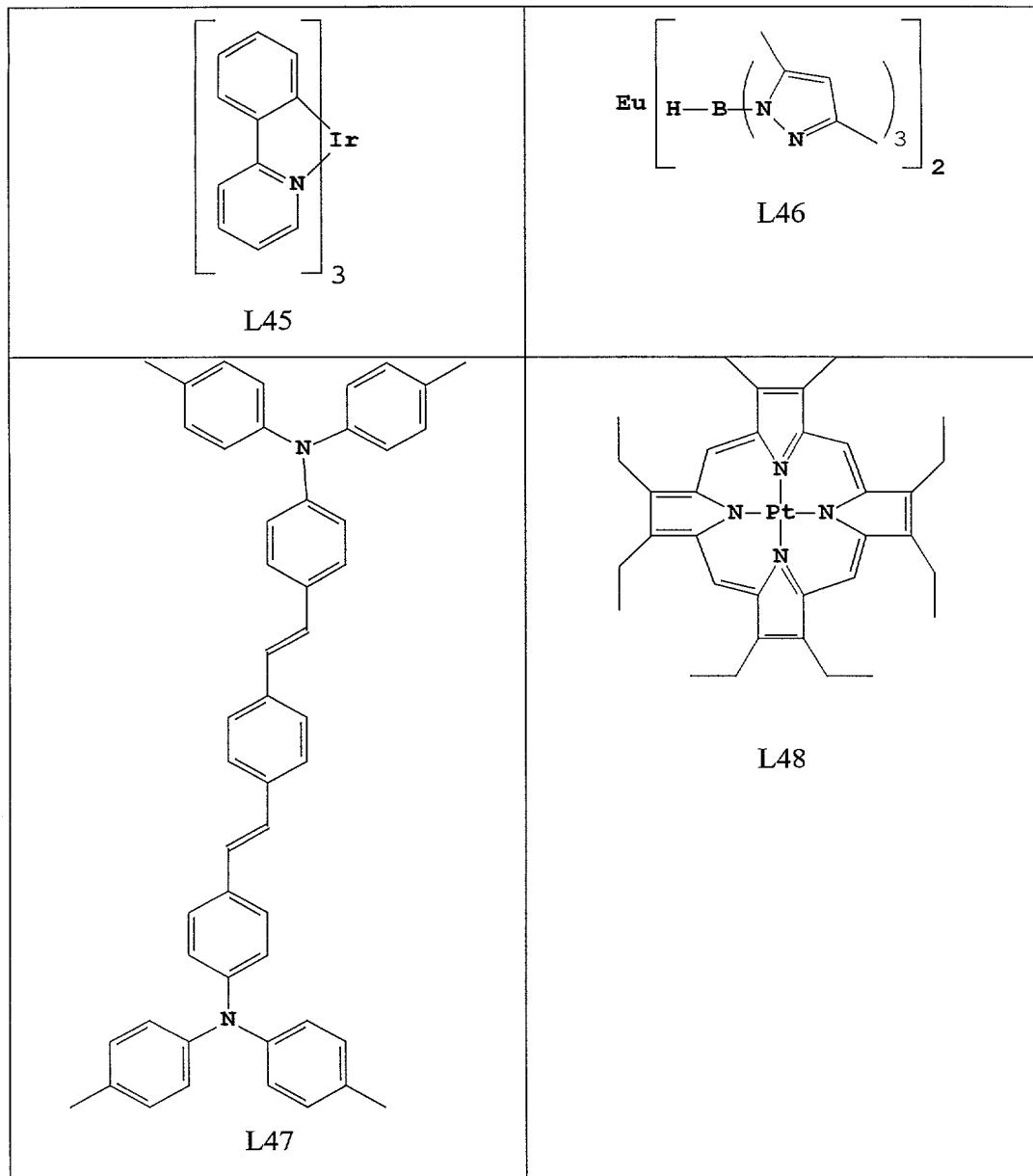
An example of a useful benzazole is 2, 2', 2''-(1,3,5-  
15 phenylene)tris[1-phenyl-1H-benzimidazole].

Distyrylarylene derivatives as described in US 5,121,029 are also useful host materials in the LEL.

Desirable fluorescent dopants include groups derived from fused  
20 ring, heterocyclic and other compounds such as anthracene, tetracene, xanthene, perylene, rubrene, coumarin, rhodamine, quinacridone, dicyanomethylenepyran, thiopyran, polymethine, pyrilium thiapyrilium, and carbostyryl compounds. Illustrative examples of useful dopants include, but are not limited to, the following:







Electron-Transporting Layer (ETL)

Preferred thin film-forming materials for use in forming the electron-transporting layer **111** of the organic EL devices of this invention are 5 metal chelated oxinoid compounds, including chelates of oxine itself (also commonly referred to as 8-quinolinol or 8-hydroxyquinoline). Such compounds help to inject and transport electrons and exhibit both high levels of performance and are readily fabricated in the form of thin films. Exemplary of contemplated

oxinoid compounds are those satisfying structural formula (E), previously described.

Other electron-transporting materials include various butadiene derivatives as disclosed in US 4,356,429 and various heterocyclic optical brighteners as described in US 4,539,507. Benzazoles satisfying structural formula (G) are also useful electron transporting materials.

In some instances, layers **109** and **111** can optionally be collapsed into a single layer that serves the function of supporting both light emission and electron transportation.

10

### Cathode

When light emission is through the anode, the cathode layer **113** used in this invention can be comprised of nearly any conductive material.

Desirable materials have good film-forming properties to ensure good contact with the underlying organic layer, promote electron injection at low voltage, and have good stability. Useful cathode materials often contain a low work function metal (< 4.0 eV) or metal alloy. One preferred cathode material is comprised of a Mg:Ag alloy wherein the percentage of silver is in the range of 1 to 20 %, as described in US 4,885,221. Another suitable class of cathode materials includes bilayers comprised of a thin layer of a low work function metal or metal salt capped with a thicker layer of conductive metal. One such cathode is comprised of a thin layer of LiF followed by a thicker layer of Al as described in US 5,677,572. Other useful cathode materials include, but are not limited to, those disclosed in US 5,059,861, US 5,059,862, and US 6,140,763.

When light emission is viewed through the cathode, the cathode must be transparent or nearly transparent. For such applications, metals must be thin or one must use transparent conductive oxides, or a combination of these materials. Optically transparent cathodes have been described in more detail in US 5,776,623. Cathode materials can be deposited by evaporation, sputtering, or chemical vapor deposition. When needed, patterning can be achieved through many well known methods including, but not limited to, through-mask deposition,

integral shadow masking as described in US 5,276,380 and EP 0 732 868, laser ablation, and selective chemical vapor deposition.

Deposition of organic layers

5        The organic materials mentioned above are suitably deposited through sublimation, but can be deposited from a solvent with an optional binder to improve film formation. If the material is a polymer, solvent deposition is usually preferred. The material to be deposited by sublimation can be vaporized from a sublimator “boat” often comprised of a tantalum material, e.g., as  
10      described in US 6,237,529, or can be first coated onto a donor sheet and then sublimed in closer proximity to the substrate. Layers with a mixture of materials can utilize separate sublimator boats or the materials can be pre-mixed and coated from a single boat or donor sheet. Patterned deposition can be achieved using shadow masks, integral shadow masks (US 5,294,870), spatially-defined thermal  
15      dye transfer from a donor sheet (US 5,851,709 and US 6,066,357) and inkjet method (US 6,066,357).

Encapsulation

20      Most OLED devices are sensitive to moisture and/or oxygen so they are commonly sealed in an inert atmosphere such as nitrogen or argon, along with a desiccant such as alumina, bauxite, calcium sulfate, clays, silica gel, zeolites, alkaline metal oxides, alkaline earth metal oxides, sulfates, or metal halides and perchlorates. Methods for encapsulation and desiccation include, but are not limited to, those described in US 6,226,890.

25

The entire contents of the patents and other publications referred to in this specification are incorporated herein by reference.

**EXAMPLES**

30      The inventions and its advantages are further illustrated by the specific examples which follow.

## Synthesis

### Example 1

Preparation of 2-Quinolylacetonitrile: To a solution of 2-(chloromethyl)quinoline (20.74 g, 117 mmol) in ethanol and water (200 mL, 4:1 ratio) was added NaCN

5 (5.73 g, 117 mmol), and the reaction mixture was heated at reflux for 18 h. The reaction mixture was cooled to ambient temperature; diluted with a mixture of heptane, ethyl acetate, and THF; and washed with a 5% aqueous solution of  $K_2CO_3$  and brine. The organic layer was isolated and dried over  $MgSO_4$ .

Purification by column chromatography with an elution gradient of 75:25

10 heptane/ethyl acetate to 65:35 heptane/ethyl acetate afforded 14.6 g (74.5%) of desired product. Results of  $^1H$  NMR spectroscopy and electrospray mass spectroscopy are consistent with the product.  $^1H$  NMR (300 MHz,  $CDCl_3$ ):  $\delta$  4.12 (s, 2H), 7.66 (m, 2H), 7.76 (m, 2H), 7.84 (d,  $J$  = 8.2 Hz, 1H), 8.06 (d,  $J$  = 8.2 Hz, 1H), 8.22 (d,  $J$  = 8.5 Hz, 1H).

15

Preparation of Bis(2-quinolyl)acetonitrile: To a solution of 2-quinolylacetonitrile (14.5 g, 86.2 mmol) in toluene (200 mL) was added slowly NaH (6.9 g of 60% oil dispersion, 172 mmol). The reaction mixture was stirred at ambient temperature for 15 min at which point  $H_2$  evolution was no longer

20 evident. A solution of 2-chloroquinoline (14.1 g, 86.2 mmol) in toluene (150 mL) was added to the reaction flask, and the reaction mixture was then heated at reflux for 18 h. The reaction mixture was cooled to ambient temperature, diluted with THF, and quenched with  $H_2O$ . The organic solution was washed with 1N HCl (200 mL), saturated aqueous solution of  $NaHCO_3$ , and brine. An orange solid 25 precipitated from the organic layer and was isolated via vacuum filtration. The mother liquor was dried over  $MgSO_4$  and the volatile components were removed with a rotary evaporator. The resulting solid was combined with the orange powder isolated via filtration. The solid was washed with a mixture of ether and heptane to afford 14.7 g (57.6%) of product. Results of  $^1H$  NMR spectroscopy and 30 electrospray mass spectroscopy are consistent with the product.  $^1H$  NMR (300 MHz,  $CDCl_3$ ):  $\delta$  7.36 (m, 2H), 7.57 (d,  $J$  = 9.1 Hz, 2H), 7.64 (m, 4H), 7.73 (d,  $J$  = 8.8 Hz, 2H), 7.86 (d,  $J$  = 9.1 Hz, 2H).

Preparation of Bis(2-quinoliny)methane: Bis(2-quinoliny)acetonitrile (14.3 g, 48.4 mmol) was dissolved in 70% H<sub>2</sub>SO<sub>4</sub> in H<sub>2</sub>O (85 mL), and the resulting solution was heated at reflux for 1.5 h. The reaction mixture was poured into a 2-L beaker full of ice and was neutralized very carefully with a NaOH solution. The aqueous solution was extracted with a mixture of ether and THF, and the organic layer was isolated and dried over MgSO<sub>4</sub>. The volatile components were removed, and the resulting solid was washed with a solution of heptane and ether to afford 11.7 g (89.5%) of bis(2-quinoliny)methane as a reddish powder. Results of <sup>1</sup>H NMR spectroscopy and electrospray mass spectroscopy are consistent with the product. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): δ 4.74 (s, 2H), 7.42 (d, *J* = 8.5 Hz, 2H), m, 2H), 7.51 (m, 2H), 7.72 (m, 2H), 7.77 (d, *J* = 8.2 Hz, 2H), 8.04 (d, *J* = 8.5 Hz, 2H), 8.11 (d, *J* = 8.5 Hz, 2H).

15 Preparation of Difluoro[1,2-dihydro-2[(2-quinoliny-κ*N*)methylene]pyridinato-κ*N*]boron (**Inv-1**): A mixture of bis(2-quinoliny)methane, diisoprylethylamine, BF<sub>3</sub> etherate, and CH<sub>2</sub>Cl<sub>2</sub> were heated at reflux for 5 h and then dilute with a solution of THF and diethyl ether. The organic solution was washed with H<sub>2</sub>O and brine, and then dried over MgSO<sub>4</sub>. The volatile components were removed with a rotary evaporator. The resulting red solid was suspended in hot ethanol and isolated via vacuum filtration. Water was added to the mother liquor to precipitate out more product, which was isolated via filtration. The combined precipitates were dried under dynamic vacuum resulting in 2.35 g (57%) of product. This product may be further purified by sublimation (165 °C @ 710 millitorr) with a N<sub>2</sub> carrier gas.

20 Results of <sup>1</sup>H NMR spectroscopy and electrospray mass spectroscopy are consistent with the product. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>): δ 5.49 (s, 1H), 6.85 (d, *J* = 9.1 Hz, 2H), 7.29 (m, 2H), 7.51 (m, 2H), 7.60 (m, 4H), 8.57 (m, 2H).

25

Example 2 – EL Device Fabrication – Inventive Example

30 An EL device satisfying the requirements of the invention was constructed as Sample 1 in the following manner:

A glass substrate coated with an 85 nm layer of indium-tin oxide (ITO) as the anode was sequentially ultrasonicated in a commercial detergent, rinsed in deionized water, degreased in toluene vapor and exposed to oxygen plasma for about 1 min.

5           a) Over the ITO was deposited a 1 nm fluorocarbon (CF<sub>x</sub>) hole-injecting layer (HIL) by plasma-assisted deposition of CHF<sub>3</sub>.

          b) A hole-transporting layer (HTL) of *N,N'*-di-1-naphthalenyl-*N,N'*-diphenyl-4, 4'-diaminobiphenyl (NPB) having a thickness of 75 nm was then evaporated from a tantalum boat.

10           c) A 25 nm light-emitting layer (LEL) of 2-*tert*-butyl-9,10-di-(2-naphthyl)anthracene (TBADN) and **Inv-1** (0.5% wt%) were then deposited onto the hole-transporting layer. These materials were also evaporated from tantalum boats.

          d) A 35 nm electron-transporting layer (ETL) of tris(8-quinolinolato)aluminum (III) (AlQ<sub>3</sub>) was then deposited onto the light-emitting layer. This material was also evaporated from a tantalum boat.

15           e) On top of the AlQ<sub>3</sub> layer was deposited a 220 nm cathode formed of a 10:1 volume ratio of Mg and Ag.

The above sequence completed the deposition of the EL device. The  
20 device was then hermetically packaged in a dry glove box for protection against ambient environment.

Samples 2 and 3 were EL devices incorporating **Inv-4** or **Inv-5** fabricated in an identical manner as the example incorporating **Inv-1**. Sample 4 is a comparative device that was prepared omitting **Inv-1** from the LEL and  
25 maintaining the same device architecture as in the inventive examples. The cells thus formed were tested for efficiency (in the form of luminance yield), and the results are listed in Table 1.

**Table 1. Evaluation Results for EL devices.**

Sample	Host	Dopant	Efficiency (cd/A) <sup>1</sup>	Type
1	TBADN	Inv-1	9.10	Invention
2	"	Inv-4	6.36	Invention
3	"	Inv-5	3.75	Invention
<b>Avg</b>			<b>6.4</b>	
4	"	None	1.41	Comparison

<sup>1</sup> Luminance yields reported at 20 mA/cm<sup>2</sup>.

As can be seen from Table 1, all tested EL devices incorporating the INV 5 dopants demonstrated superior luminance yields relative to the comparative device containing only TBADN. These doped EL devices exhibit green electroluminescence with  $\lambda_{max}$  ranging from 508-532 nm. In contrast, blue emission ( $\lambda_{max}$  of 448 nm), attributed to TBADN fluorescence, is generated in the light-emitting layer of comparative Sample 4.

10 In addition to the high luminance yields demonstrated by EL devices containing bis(azinyl)methene boron complexes, the stability of these EL devices also appears to be increased over that of a device without that material. For example, a series of EL devices (Samples 5-9) containing **Inv-1** and TBADN and a comparative device (Sample 10) without **Inv-1** were prepared and subjected to a 15 constant current density of 20 mA/cm<sup>2</sup> at 70 °C. The percent loss of initial luminance was measured after 300 hours, and the results are reported in Table 2. As can be seen from the data, the EL devices containing **Inv-1** retain far more of their initial luminance intensity than did the comparative example, with an average improvement of about 35%.

**Table 2. EL Device Stability Comprised of Inv-1 in TBADN**

Sample	Type	Dopant wt%	luminance loss - % <sup>1</sup>
5	Invention	(0.5%)	28.8
6	Invention	(1.0%)	28.1
7	Invention	(1.5%)	31.6
8	Invention	(2.0%)	32.1
9	Invention	(2.5%)	36.2
<b>Avg.</b>			<b>31.4</b>
10	Comparison	(0.0%)	43.6

<sup>1</sup> Percent loss of initial luminance after 300 hours at constant current density (20 mA/cm<sup>2</sup>) and temperature (70 °C).

5    Example 3 – EL Device Fabrication – Inventive Example

An EL device satisfying the requirements of the invention was constructed as Sample 11 in the following manner:

10    A glass substrate coated with an 85 nm layer of indium-tin oxide (ITO) as the anode was sequentially ultrasonicated in a commercial detergent, rinsed in deionized water, degreased in toluene vapor and exposed to oxygen plasma for about 1 min.

- a) Over the ITO was deposited a 1 nm fluorocarbon (CFx) HIL by plasma-assisted deposition of CHF<sub>3</sub>.
- b) A HTL of *N,N'*-di-1-naphthalenyl-*N,N'*-diphenyl-4, 4'-diaminobiphenyl (NPB) having a thickness of 75 nm was then evaporated from a tantalum boat.
- c) A 37.5 nm LEL of tris(8-quinolinolato)aluminum (III) (AlQ<sub>3</sub>) and Inv-1 (0.4 wt%) were then deposited onto the hole-transporting layer. These materials were also evaporated from tantalum boats.
- d) A 37.5 nm ETL of tris(8-quinolinolato)aluminum (III) (AlQ<sub>3</sub>) was then deposited onto the light-emitting layer. This material was also evaporated from a tantalum boat.
- e) On top of the AlQ<sub>3</sub> layer was deposited a 220 nm cathode formed of a 10:1 volume ratio of Mg and Ag.

25    The above sequence completed the deposition of the EL device. The device was then hermetically packaged in a dry glove box for protection against ambient environment.

A comparative device was prepared as Sample 12 omitting **Inv-1** from the light-emitting layer and maintaining the same device architecture as this example. The cells thus formed were tested for efficiency (in the form of luminance yield), and the results are listed in Table 3.

5

**Table 3. Evaluation Results for EL devices.**

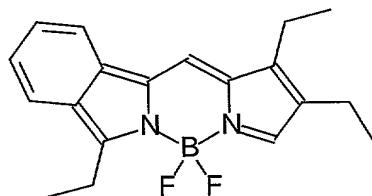
Sample	Host	Dopant	Efficiency (cd/A) <sup>1</sup>	Type
11	AlQ <sub>3</sub>	<b>Inv-1</b>	10.39	Invention
12	"	None	3.40	Comparison

<sup>1</sup> Luminance yields reported at 20 mA/cm<sup>2</sup>

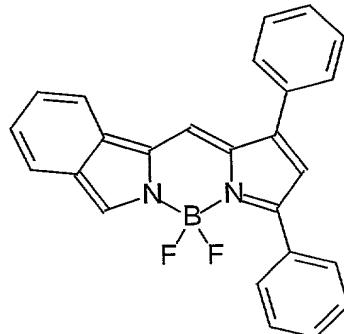
As can be seen from Table 3, the tested EL device incorporating the INV 10 dopant in an AlQ<sub>3</sub> host demonstrates a superior luminance yield relative to the comparative device. The inventive device exhibited green emission ( $\lambda_{\text{max}}$  of 540 nm) with a well-defined vibronic structure and narrow emission peaks as opposed to the very broad AlQ<sub>3</sub> emission at 540 nm.

EL data for the 5,6,5-tricyclic pyrromethene BF<sub>2</sub> complexes, presented in 15 patents US 5,683,823, JP 09,289,081A, and JP 11,097,180A is not directly comparable with the EL data obtained above. Such compounds are exemplified by the following:

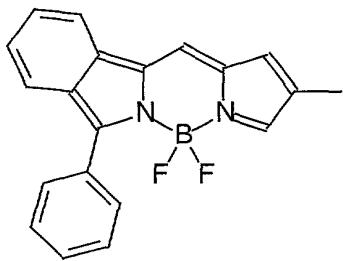
**Comp-1**



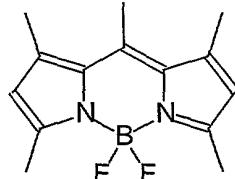
**Comp-2**



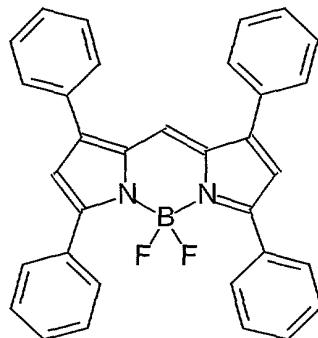
Comp-3



Comp-4



Comp-5



The reported luminance yields for the 5,6,5-tricyclic pyrromethene  $\text{BF}_2$  complexes appear to be lower on average than those yields of the inventive examples tested. In addition, of the 5,6,5-tricyclic pyrromethene  $\text{BF}_2$  complexes reported, only 4,4-difluoro-1,3,5,7,8-pentamethyl-4-bora-3a,4a-diaza-s-indacene 5 exhibited green electroluminescence at 2.58 cd/A as reported in JP 11,097,180A. All other EL devices containing 5,6,5-tricyclic pyrromethene  $\text{BF}_2$  complexes exhibited red electroluminescence. It appears that the bis(azinyl)methene boron group is a necessary component to generate highly efficient EL devices with green emission and desirable stability.

**PARTS LIST**

- 101 Substrate
- 103 Anode
- 105 Hole-Injecting layer (HIL)
- 107 Hole-Transporting layer (HTL)
- 109 Light-Emitting layer (LEL)
- 111 Electron-Transporting layer (ETL)
- 113 Cathode